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CLAIMS

1 Claim 1. In a process for producing an olefin by the vapor
2 phase oxidative dehydrogenation of alkane having from 2 to 5
3 carbon atoms in the presence of molecular oxygen, the
4 improvement comprising, carrying out the vapor phase oxidative
5 dehydrogenation in the presence of an oxidative dehydrogenation
6 catalyst comprising a multimetal mixed oxide having the formula



8 where Mn denotes manganese;

9 O denotes oxygen;

10 E¹ represents one or more metal elements selected from the
11 group consisting of phosphorus, arsenic, antimony, boron, sulfur,
12 selenium, tellurium, fluorine, chlorine, bromine, iodine, niobium,
13 tantalum, tungsten, rhenium and copper;

14 E² represents one or more metal elements selected from the
15 group consisting of chromium, iron, cobalt, nickel, silver, gold, zinc,
16 thallium, tin, lead, bismuth, lithium, sodium, potassium, rubidium,
17 cesium, magnesium, calcium, strontium, barium, yttrium, lanthanum,
18 cerium, neodymium, and samarium; and,

19 α, β, γ and x denote atomic numbers of Mn, E¹, E², and oxygen,
20 respectively, and,

21 when α = 1, β = 0.01-10, γ = 0-5, and x has a numerical value
22 determined by the state of oxidation of the elements other than
23 oxygen.

1 Claim 2. The process according to claim 1, wherein, in the
2 oxidative dehydrogenation catalyst of formula (1), when α = 1,
3 β = 0.02-2 and γ = 0-1.

1 Claim 3. The process according to claim 2, wherein E¹
2 comprises at least sulfur, and the sulfur is added in the form of its
3 sulfate ion (SO₄²⁻).

1 Claim 4. The process according to claim 1, wherein E¹
2 comprises at least sulfur, and the sulfur is added in the form of its
3 sulfate ion (SO₄²⁻).

1 Claim 5. The process according to claim 1, wherein the
2 oxidative dehydrogenation catalyst of formula (1) is one which is
3 dried and fired at temperatures not higher than 300°C.

1 Claim 6. The process according to claim 1, wherein the
2 oxidative dehydrogenation catalyst is supported on a refractory
3 inorganic carrier.

1 Claim 7. The process according to claim 1, wherein the
2 oxidative dehydrogenation of said alkane is carried out at a space
3 velocity of from 300 to 30,000 hr⁻¹, and at a temperature of from 250
4 to 650 °C.

1 Claim 8. The process according to claim 1, wherein the
2 oxidative dehydrogenation catalyst is selected from the group
3 consisting of

4 $\text{Mn}_1\text{Sb}_{0.15}\text{O}_x$, $\text{Mn}_1\text{Sb}_{0.25}\text{O}_x$, $\text{Mn}_1\text{B}_{0.1}\text{O}_x$, $\text{Mn}_1\text{S}_{0.1}\text{O}_x$, $\text{Mn}_1\text{Nb}_{0.05}\text{O}_x$, $\text{Mn}_1\text{W}_{0.05}\text{O}_x$,
 5 $\text{Mn}_1\text{Re}_{0.05}\text{O}_x$, $\text{Mn}_1\text{Cu}_{0.1}\text{O}_x$, $\text{Mn}_1\text{Cl}_{0.4}\text{O}_x$, $\text{Mn}_1\text{Sb}_{0.15}\text{Cr}_{0.1}\text{O}_x$, $\text{Mn}_1\text{Sb}_{0.15}\text{Na}_{0.1}\text{O}_x$,
 6 $\text{Mn}_1\text{Sb}_{0.15}\text{Mg}_{0.1}\text{O}_x$, $\text{Mn}_1\text{Sb}_{0.15}\text{Ce}_{0.1}\text{O}_x$, $\text{Mn}_1\text{S}_{0.15}\text{Cr}_{0.1}\text{O}_x$, $\text{Mn}_1\text{Cl}_{0.4}\text{Sn}_{0.1}\text{O}_x$,
 7 $\text{Mn}_1\text{Sb}_{0.15}\text{W}_{0.05}\text{Cr}_{0.1}\text{O}_x$, $\text{Mn}_1\text{Sb}_{0.15}\text{W}_{0.05}\text{S}_{0.15}\text{Cr}_{0.1}\text{O}_x$, and
 8 $\text{Mn}_1\text{Sb}_{0.15}\text{W}_{0.05}\text{Nb}_{0.05}\text{Cr}_{0.1}\text{O}_x$.

1 Claim 9. In a process for producing unsaturated aldehyde and
2 unsaturated acid by the vapor phase oxidative dehydrogenation of
3 C₂ to C₅ alkene in the presence of molecular oxygen, the
4 improvement comprising, carrying out the vapor phase oxidative

5 dehydrogenation in the presence of an oxidative dehydrogenation
6 catalyst comprising a multimetal mixed oxide having the formula



8 where Mn denotes manganese;

9 O denotes oxygen;

10 E¹ represents one or more metal elements selected from the
11 group consisting of phosphorus, arsenic, antimony, boron, sulfur,
12 selenium, tellurium, fluorine, chlorine, bromine, iodine, niobium,
13 tantalum, tungsten, rhenium and copper;

14 E² represents one or more metal elements selected from the
15 group consisting of chromium, iron, cobalt, nickel, silver, gold, zinc,
16 thallium, tin, lead, bismuth, lithium, sodium, potassium, rubidium,
17 cesium, magnesium, calcium, strontium, barium, yttrium, lanthanum,
18 cerium, neodymium, and samarium; and,

19 α , β , γ and x denote atomic numbers of Mn, E¹, E², and oxygen,
20 respectively, and,

21 when $\alpha = 1$, $\beta = 0.01-10$, $\gamma = 0-5$, and x has a numerical value
22 determined by the state of oxidation of the elements other than
23 oxygen.

1 Claim 10. The process according to claim 9, wherein, in the
2 oxidative dehydrogenation catalyst of formula (1), when $\alpha = 1$, $\beta =$
3 0.02-2 and $\gamma = 0-1$.

1 Claim 11. The process according to claim 10, wherein E¹
2 comprises at least sulfur, and the sulfur is added in the form of its
3 sulfate ion (SO₄²⁻).

1 Claim 12. The process according to claim 9, wherein E¹
2 comprises at least sulfur, and the sulfur is added in the form of its
3 sulfate ion (SO₄²⁻).

1 Claim 13. The process according to claim 9, wherein the
2 oxidative dehydrogenation catalyst of formula (1) is one which is
3 dried and fired at temperatures not higher than 300°C.

1 Claim 14. The process according to claim 9, wherein the
2 oxidative dehydrogenation catalyst is supported on a refractory
3 inorganic carrier.

4 Claim 15. The process according to claim 9, wherein the
5 oxidative dehydrogenation of said alkene is carried out at a space
6 velocity of from 300 to 30,000 hr⁻¹, and at a temperature of from
7 250 to 650 °C.

8 / \Rightarrow Claim 16. The process according to claim 9, wherein the
9 oxidative dehydrogenation catalyst of formula (1) is a catalyst of
10 the following formula (2):

11 $\text{Mo}_a\text{Bi}_b\text{Fe}_c\text{A}_d\text{B}_e\text{C}_f\text{D}_g\text{O}_x$ (2),

12 wherein Mo denotes molybdenum, Bi denotes bismuth, Fe
13 denotes iron, O denotes oxygen,

14 A represents at least one metal element selected from the
15 group consisting of cobalt and nickel,

16 B represents at least one metal element selected from the
17 group consisting of alkali metals and thallium,

18 C represents at least one metal element selected from the
19 group consisting of silicon, aluminum, zirconium and titanium,

20 D represents at least one metal element selected from the
21 group consisting of tungsten, phosphorus, tellurium, antimony,
22 tin, cerium, lead, niobium, manganese, arsenic and zinc,

23 a, b, c, d, e, f, g, and x represent the atomic ratios of the
24 respective elements, and, when a = 12, b = 0.1-10, c = 0.1-20, d = 2-20,
25 e = 0.001 = 10, f = 0-30, g = 0-4, and x is a numerical value
26 determined by the state of oxidation of the elements other than
27 oxygen.

1 1 Claim 17. In a process for producing unsaturated acid having
2 from 2 to 5 carbon atoms, by the vapor phase oxidative
3 dehydrogenation of the corresponding unsaturated aldehyde of
4 from 2 to 5 carbon atoms, in the presence of molecular oxygen,
5 the improvement comprising, carrying out the vapor phase
6 oxidative dehydrogenation in the presence of an oxidative
7 dehydrogenation catalyst comprising a multimetal mixed oxide
8 having the formula

9 $Mn_{\alpha}E_1^{\beta}E_2^{\gamma}Ox$ (1)

10 where Mn denotes manganese;

11 O denotes oxygen;

12 E¹ represents one or more metal elements selected from the
13 group consisting of phosphorus, arsenic, antimony, boron, sulfur,
14 selenium, tellurium, fluorine, chlorine, bromine, iodine, niobium,
15 tantalum, tungsten, rhenium and copper;

16 E² represents one or more metal elements selected from the
17 group consisting of chromium, iron, cobalt, nickel, silver, gold,
18 zinc, thallium, tin, lead, bismuth, lithium, sodium, potassium,
19 rubidium, cesium, magnesium, calcium, strontium, barium
20 yttrium, lanthanum, cerium, neodymium, and samarium; and,

21 α , β , γ and x denote atomic numbers of Mn, E¹, E², and
22 oxygen, respectively, and,

23 when $\alpha = 1$, $\beta = 0.01-10$, $\gamma = 0-5$, and x has a numerical value
24 determined by the state of oxidation of the elements other than
25 oxygen.

1 Claim 18. The process according to claim 17, wherein the
2 oxidative dehydrogenation catalyst is represented by the
3 following formula (3):

$$4 \quad \text{Mo}_p\text{V}_i\text{W}_j\text{E}_k\text{F}_l\text{G}_m\text{H}_n\text{O}_x \quad (3)$$

5 where Mo denotes molybdenum, V denotes vanadium, W
 6 denotes tungsten, O denotes oxygen,

7 E represents at least one element selected from the group
8 consisting of copper, cobalt, bismuth and iron,

9 F represents at least one element selected from the group
10 consisting of antimony and niobium,

11 G represents at least one element selected from the group
12 consisting of silicon, aluminum, zirconium, and titanium,

13 H represents at least one element selected from the group
14 consisting of alkaline earth metals, thallium, phosphorus,
15 tellurium, tin, cerium, lead, manganese and zinc;

16 h, i, j, k, l, m, n, and x represent the atomic ratios of the
17 respective elements, and, when h=12, i=0.1-10, j=0-10, k=0.1-20,
18 l=0-10, m=0-10, n=0-30, and x has a numerical value determined
19 by the state of oxidation of the elements other than oxygen.

1 > Claim 19. A process for producing unsaturated acid by the
2 vapor phase oxidative dehydrogenation of C₂ to C₅ alkene in the
3 presence of molecular oxygen, which comprises, oxidatively
4 dehydrogenating lower alkene of from 2 to 5 carbon atoms in the
5 vapor phase in the presence of oxidative dehydrogenation
6 catalyst comprising a multimetal mixed oxide having the formula



8 where Mn denotes manganese;

9 O denotes oxygen;

10 E¹ represents one or more metal elements selected from the
11 group consisting of phosphorus, arsenic, antimony, boron, sulfur,
12 selenium, tellurium, fluorine, chlorine, bromine, iodine, niobium,
13 tantalum, tungsten, rhenium and copper;

14 E² represents one or more metal elements selected from the
15 group consisting of chromium, iron, cobalt, nickel, silver, gold,
16 zinc, thallium, tin, lead, bismuth, lithium, sodium, potassium,
17 rubidium, cesium, magnesium, calcium, strontium, barium
18 yttrium, lanthanum, cerium, neodymium, and samarium; and,

19 α , β , γ and x denote atomic numbers of Mn, E¹, E², and
20 oxygen, respectively, and,

21 when $\alpha = 1$, $\beta = 0.01-10$, $\gamma = 0-5$, and x has a numerical value
22 determined by the state of oxidation of the elements other than
23 oxygen, to produce a mixture of unsaturated aldehyde and
24 unsaturated acid, and

subjecting the unsaturated aldehyde to further vapor phase oxidative dehydrogenation in the presence of oxidative dehydrogenation catalyst of formula (1) to produce the corresponding unsaturated acid.

1 Claim 20. The process according to claim 19, wherein the
2 oxidative dehydrogenation catalyst used in the oxidative
3 dehydrogenation of said unsaturated aldehyde is represented by
4 the following formula (3):



6 where Mo denotes molybdenum, V denotes vanadium, W
7 denotes tungsten, O denotes oxygen,

8 E represents at least one element selected from the group
9 consisting of copper, cobalt, bismuth and iron,

10 F represents at least one element selected from the group
11 consisting of antimony and niobium,

12 G represents at least one element selected from the group
13 consisting of silicon, aluminum, zirconium, and titanium,

14 H represents at least one element selected from the group
15 consisting of alkaline earth metals, thallium, phosphorus,
16 tellurium, tin, cerium, lead, manganese and zinc;

17 h, i, j, k, l, m, n, and x represent the atomic ratios of the
18 respective elements, and, when h = 12, i = 0.1-10, j = 0-10, k = 0.1-20,
19 l = 0-10, m = 0-10, n = 0-30, and x has a numerical value determined
20 by the state of oxidation of the elements other than oxygen.

1 Claim 21. The process according to claim 20, wherein the
2 oxidative dehydrogenation catalyst used for the oxidative
3 dehydrogenation of said alkene to produce said mixture is a
4 catalyst of the following formula (2):

$$5 \quad \text{Mo}_a\text{Bi}_b\text{Fe}_c\text{A}_d\text{B}_e\text{C}_f\text{D}_g\text{O}_x \quad (2),$$

6 wherein Mo denotes molybdenum, Bi denotes bismuth, Fe
7 denotes iron, O denotes oxygen,

8 A represents at least one metal element selected from the
9 group consisting of cobalt and nickel,

10 B represents at least one metal element selected from the
11 group consisting of alkali metals and thallium,

12 C represents at least one metal element selected from the
13 group consisting of silicon, aluminum, zirconium and titanium,

14 D represents at least one metal element selected from the
15 group consisting of tungsten, phosphorus, tellurium, antimony,
16 tin, cerium, lead, niobium, manganese, arsenic and zinc,

17 a, b, c, d, e, f, g, and x represent the atomic ratios of the
18 respective elements, and, when a = 12, b = 0.1-10, c = 0.1-20, d = 2-20,
19 e = 0.001-10, f = 0-30, g = 0-4, and x is a numerical value
20 determined by the state of oxidation of the elements other than
21 oxygen

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